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(71) Applicant: TORAY INDUSTRIES, INC.  
2, Nihonbashi-Muromachi 2-chome Chuo-ku  
Tokyo 103(JP)

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(72) Inventor: Yamamoto, Yoshiyuki  
1490, Ikeda-cho  
Suzuka-shi Mie-ken(JP)

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(72) Inventor: Inoue, Toshihide  
23-40, Aza Miyazakihishi Tozuka Yamato-cho  
Ichinomiya-shi Aichi-ken(JP)

(72) Inventor: Yanagi, Masana  
6-70-2, Maruya-cho Showa-ku  
Nagoya-shi Aichi-ken(JP)

(74) Representative: Dres. Kador & Klunker  
Corneliusstrasse 15  
D-8000 München 5(DE)

(52) Flame retarding thermoplastic polyester composition.

(57) A flame retarding thermoplastic polyester composition superior in thermal stability, fluidity and mechanical properties, and prepared by mixing (a) halogen containing polystyrene and/or halogen containing poly- $\alpha$ -methylstyrene, (b) antimony trioxide and (c) epoxy compound with respect to thermoplastic polyester is disclosed.

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Title of the Invention

Flame retarding thermoplastic polyester composition

Background of the Invention

Field of the Invention:

The present invention relates to a flame retarding polyester composition superior in thermal stability, fluidity and mechanical properties.

Description of the Prior Art:

Thermoplastic polyesters such as polyethylene terephthalate, polybutylene terephthalate, polyethylene naphthalate, polycyclohexane dimethylene terephthalate, etc. have found applications in mechanical components, electrical parts, motor vehicle parts and the like. On the other hand, for industrial materials as described above, safety against flames, i.e. flame retardancy, has been strongly required. Besides balancing general chemical and physical properties, with the further requirement that, during injection molding of molded items or heat treatment thereof, the flame retarding agent does not bleed out onto the surfaces of the molded items.

Conventionally, as non-bleed type flame retarding agents for the thermoplastic polyester, there have been proposed polymeric flame retarding agents such as a halogenated polycarbonate (Japanese Laid Open Patent

Application Tokkaisho No. 48-52834) and a halogenated epoxy resin (Japanese Patent Publication Tokkosho No. 53-18068), etc. Although the known flame retarding agents as described above, which are mainly composed of tetrabromobisphenol A, are superior in compatibility with respect to polyester, they tend to deteriorate the crystallinity inherent in polyester, thus adversely affecting the mechanical properties, thermal stability, chemical resistance, and electrical properties, etc. of the polyester itself.

Meanwhile, there have also been introduced halogen containing polystyrene and halogen containing poly- $\alpha$ -methylstyrene, together with their applications to thermoplastic polyesters (Plastic Technology, July, 1980, pages 71 -- 74), but in the polyester compounds in which the above flame retarding agents are mixed, marked deterioration of thermal stability and mechanical properties is noticed, with simultaneous reduction of fluidity, thus resulting in poor moldability.

Therefore, it has been desired to realize polyester compounds free from undesirable bleeding out of the flame retarding agent, with simultaneous superior flame retardancy and favorable mechanical properties of polyester itself.

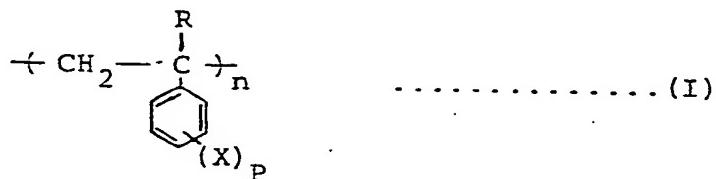
## Summary of the Invention

Accordingly, an object of the present invention is to provide a flame retarding polyester compound which is superior in balancing of thermal stability, fluidity and mechanical properties, without bleeding-out of the flame retarding agent.

It has been found that, by employing halogen containing polystyrene and/or halogen containing poly- $\alpha$ -methylstyrene as flame retarding agents, with a further addition thereto of epoxy compounds, a flame retarding polyester compound having superior flame retardancy and also improved thermal stability and fluidity, and yet favorably maintaining the mechanical properties of polyester itself, can be obtained.

More specifically, the present invention provides a flame retarding polyester composition which comprises

(a) 3 to 45 parts by weight of halogen containing polystyrene and/or halogen containing poly- $\alpha$ -methylstyrene represented by the general formula (I) given below, (b) 1 to 20 parts by weight of antimony trioxide and (c) 0.1 to 10 parts by weight of epoxy compound which are mixed to 100 parts by weight of thermoplastic polyester.



(wherein R is a hydrogen atom or methyl group, X is a

bromine or chlorine atom, P is an integer of 1 to 5, and n represents an integer larger than 2).

Description of the Preferred Embodiments

The main component of the thermoplastic polyesters employed in the practice of the present invention are polymers or copolymers prepared by condensing a dicarboxylic acid (or an ester forming compound thereof) with a glycol (or an ester forming compound thereof).

Among the dicarboxylic acids suitable for preparing polyesters in accordance with the present invention are the aromatic dicarboxylic acids such as terephthalic acid, isophthalic acid, orthophthalic acid, 2,6-naphthalene dicarboxylic acid, 1,5-naphthalene dicarboxylic acid, bis-(p-carboxyphenyl) methane, 1,2-bis(p-carboxyphenoxy) ethane, anthracene dicarboxylic acid, 4,4'-diphenyl dicarboxylic acid, 4,4'-diphenylether dicarboxylic acid, aliphatic dicarboxylic acids such as adipic acid, sebacic acid, azelaic acid, cycloaliphatic dicarboxylic acids such as 1,3-cyclohexane dicarboxylic acid, 1,4-cyclohexane dicarboxylic acid and the like, or ester-forming compounds thereof.

The glycols suitable for preparing polyesters in accordance with the present invention include aliphatic diols having 2 to 10 carbon atoms such as ethylene glycol, 1,3-propylene glycol, 1,4-butanediol, neopentylglycol,

1,5-pentanediol, 1,6-hexanediol, decamethylene glycol, cyclohexanediol, long chain glycols having molecular weights on the order of about 400 -- 6,000 such as poly(ethylene glycol), poly(1,3,-propylene glycol), poly(tetramethylene glycol) and the like.

Mixtures of these glycols can also be employed.

Preferable polyesters are poly(ethylene terephthalate), poly(propylene terephthalate), poly(butylene terephthalate), poly(butylene sebacate), poly(ethylene 2,6-naphthalate), and copolyesters such as poly(ethylene isophthalate-terephthalate) (PETI), poly(butylene terephthalate-isophthalate) (PBTI) and the like.

Among these poly(ethylene terephthalate) and poly(butylene terephthalate) are more preferably used because of their excellent mechanical properties.

Furthermore, mixtures as desired of the above polyesters and copolyesters may also be employed, and even a mixture of other organic polymer less than 30 parts by weight, with respect to 100 parts by weight of those polyesters can be applied to the present invention.

As the organic copolymers employable in the above case, there may be mentioned polyethylenes, polypropylenes, ethylene-vinylacetate-copolymers, ethylene-acrylic ester-copolymers, ethylene-propylene-copolymers, butyl rubber, chlorinated polyethylenes, chlorinated polypropylenes, polybutadiene, polystyrene, styrene-acrylonitrile-butadiene resins, styrene-polyphenylene oxide resins, styrene-acrylonitrile copolymers, polyvinyl chlorides, polyoxymethylenes, polyamides, polyphenylene oxides,

polytetrafluoroethylenes, ethylene tetrafluoroethylene copolymers, polycarbonates, etc.

The second component (a) to be employed as the flame retarding agent in the present invention is the halogen containing polystyrene and/or halogen containing poly- $\alpha$ -methylstyrene represented by the general formula given earlier and may be produced by subjecting the halogenated styrene or halogenated- $\alpha$ -methylstrene such as tribromostyrene to polymerization or by halogenating polystyrene or poly- $\alpha$ -methylstyrene.

Meanwhile, the flame retarding agent of the general formula (I) can be used, even if other vinyl group compounds are copolymerized therewith. The vinyl group compounds in the above case include styrene,  $\alpha$ -methylstyrene, acrylonitrile, methyl acrylate, butyl acrylate, methyl methacrylate, butadiene, vinyl acetate, etc.

The amount of addition of halogen containing polystyrene and/or halogen containing poly- $\alpha$ -methyl styrene represented by the general formula (I) is 3 to 45 parts by weight and more preferably, 5 to 40 parts by weight with respect to 100 parts by weight of thermoplastic polyester, and if it is less than 3 parts by weight, sufficient flame retardancy is not obtained, while on the contrary, if it exceeds 45 parts by weight, the mechanical properties of the thermoplastic polyester are undesirably impaired.

The flame retarding effect of the halogen containing

polystyrene and/or halogen containing poly- $\alpha$ -methyl styrene may be remarkably improved by the simultaneous addition thereto of (b) antimony trioxide for a third component as a flame retarding assistant. The amount of addition thereof should be 1 to 20 parts by weight and more preferably, 3 to 18 parts by weight with respect to 100 parts by weight of thermoplastic polyester, and if it is less than 1 part by weight, the flame retardancy is not sufficient, while, if it exceeds 20 parts by weight, the mechanical properties of the thermoplastic polyester are undesirably impaired.

Still more preferably, it is desirable to add antimony trioxide at the rate of one antimony atom for every 2 to 5 halogen atoms in the added halogen containing polystyrene and/or halogen containing poly- $\alpha$ -methystyrene. Simultaneously, other flame raterding assistants such as boron oxide, zirconium oxide, iron oxide, etc. may be employed.

Although the composition prepared by mixing the flame retarding agent represented by the formula (I) and antimony trioxide, with the thermoplastic polyester, has a superior flame retardancy as compared with the conventional flame retarding polyesters, if (c) epoxy compound is further added thereto as a further component, further improvements of mechanical properties such as impact resistance, etc., and also of thermal stability may be achieved with simultaneous improvements of fluidity stabilization of the

composition during molding in the fluid state.

As the epoxy compounds which may be employed for the present invention, there may be mentioned, for example, epoxy ethers such as  $\beta,\gamma$ -epoxypropyl ether, 1,4-bis( $\beta,\gamma$ -epoxypropyl ether), 1,4-bis( $\beta,\gamma$ -epolypropoxy) butane, 1,6-bis(epoxy ethyl) hexane, 2,2-bis[p-( $\beta,\gamma$ -epoxypropoxy)-phenyl] propane, 1-epoxyethyl-3,4-epoxycyclohexane, 1-( $\beta,\gamma$ -epoxypropoxy)-2-benzyl oxyethane, 1-( $\beta,\gamma$ -epoxypropoxy) 2-ethoxyethane, 1,4-bis( $\beta,\gamma$ -epoxypropoxy) benzene, etc., and epoxy esters and their derivatives such as tetrahydrophthalic acid diglycidyl ester, hexahydrophthalic acid diglycidyl ester, phthalic acid diglycidyl ester, dimer acid diglycidyl ester, octadecanoic dicarboxylic acid diglycidyl ester, aliphatic acid monoglycidyl ester, etc., various copolymers containing epoxy groups prepared by copolymerization of unsaturated monomers containing epoxy groups and other unsaturated monomers such as ethylene/glycidyl methacrylate copolymer, etc.

Particularly, undecanoic carboxylic acid monoglycidyl ester, tetrahydrophthalic acid diglycidyl ester, hexahydrophthalic acid diglycidyl ester, ethylene/glycidyl methacrylate copolymer, and bis-glycidyl polyether (oligomer at quantity of 1 to 10) obtained by the reaction between epichlorohydrin and 2,2-bis-(4-hydroxylphenol) propane (referred to as bis-phenol A) may be favorably employed.

The amount of addition of these epoxy compounds should be in the range of 0.1 to 10 parts by weight and more preferably, 0.2 to 9 parts by weight with respect to 100 parts by weight of the thermoplastic polyester, and if it is less than 0.1 part by weight, the improvement of mechanical properties, fluidity, etc., is small, while on the contrary, if it exceeds 10 parts by weight, the flame retardancy is undesirably impaired. If desired, to the composition of the present invention may be added various additives such as fibrous or granular reinforcements and fillers including glass fibers, glass beads, carbon fibers, calcium carbonate, magnesium carbonates, barium sulfates, granular silicas, wollastonites, talcs, clays, micas, sericitics, zeolites, bentonites, dolomites, kaolinites, asbestos, feldspars and the like; antioxidants and heat-stabilizers selected from hindered phenols, hydroquinones, thioethers, phosphites and the like; ultraviolet absorbents selected from various substituted benzotriazoles, benzophenones, salicylates, resorcinols and the like; lubricants and mold release agents selected from stearic acid, stearates, montanic acid, montan wax, montanates, stearyl alcohol, stearyl amides such as ethylene bis(stearamide) and the like; coloring agents including dyes and pigments such as carbon black, titanium dioxide, cadmium sulfide and phthalocyanines; another flame retardants selected from halogen substituted aromatic compounds such as decarbromodiphenyl ether,

brominated polycarbonates, melamine compounds, cyanuric acid derivatives, phosphorus compounds and the like; nucleating agents and antiwear agents and the like, as long as the characteristics of the resulting compositions are not adversely affected. More than two of above mentioned additives can be added. Further, if desired, to the composition of the present invention may be added small amounts of other thermoplastic resins such as polyethylenes, polypropylenes, acrylic resins, fluoro-containing polymers, polyacetals, polycarbonates, polysulfones or polyphenylene-oxides, thermosetting resins such as phenolic resins, melamine resins, unsaturated polyester resins, silicone resins or epoxy resins, and elastomeric thermoplastic resins such as ethylene-vinylacetate-copolymers, ethylene-propylene-terpolymers, block-copolyetheresters and the like. More than the two types of the above mentioned resins may be added.

The composition of the present invention may be produced by the conventional methods, which include, for example, the process in which the thermoplastic polyester, halogen containing polystyrene and/or halogen containing poly- $\alpha$ -methylstyrene, antimony trioxide, epoxy compound and other necessary additives are subjected to melting and mixing in an extruder or kneader; the process in which, after uniformly mixing granular materials with each other mechanically, molding is effected simultaneously with mixing by a direct injection molding machine; and another

process in which the additives are directly charged into a polymerizing vessel of resin for mixing, etc.

In the above case, although the addition may be effected in any order as desired, it is one of the preferable methods to mix the halogen containing polystyrene and/or halogen containing poly- $\alpha$ -methylstyrene and antimony trioxide, after subjecting the thermoplastic polyester and epoxy compound to preliminary mixing.

The resinous composition of the present invention can be molded according to conventional methods such as extrusion molding, blow molding, injection molding and the like.

The molded products as described above are not only favorable in the flame retardancy, but also superior in fluidity and thermal stability, with a good surface appearance, and therefore, useful for mechanical components, electrical parts, and parts for motor vehicles, etc.

Hereinbelow, examples are inserted for the purpose of illustrating the present invention, without any intention of limiting the scope thereof.

It is to be noted that, in the examples to be given hereinbelow, % and parts are all based on weight, and that, in the illustrative examples and comparative examples, the properties of the resinous composition are determined according to the following methods:

Relative viscosity : With o-chlorophenol employed as a solvent, polymer solution at 25 % is measured at 25 °C

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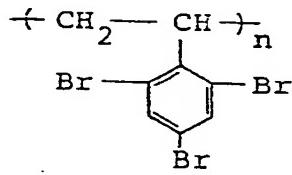
- Tensile property : ASTM D-636
- Flammability : UL-94 (vertical burning test)
- Thermal stability : Tensile strength maintaining rate (%)  
after subjecting the molded products  
to dry heat treatment at 190 °C for  
15 days
- Fluidity : Filling minimum pressure (gauge pressure)  
during injection of the tensile molding  
piece is measured.

EXAMPLE 1

Polyethylene terephthalate (PET) with a relative viscosity of 1.35, flame retarding agent (A) as stated below, glass fibers (chopped strand of 3 mm in length), antimony trioxide (SBO), and bis-glycidil polyether of bisphenol A ("EPIKOTE 815" made by Shell Company) were mixed by a V-blender at the rate shown in Table 1, and further melted for kneading at 280°C by an extruder of 65 mmØ so as to be formed into pellets. After subjecting these pellets to vacuum drying at 130°C for 5 hours, burning test pieces (1/8" x 1/2" x 5" and 1/32" x 1/2" x 5") and tensile dumbbell test pieces were molded at a molding temperature of 280°C and a mold temperature of 130°C, and the minimum injection pressure at this time was recorded.

With the use of the above burning test pieces, the vertical burning test was effected according to UL-94 standard, while the tensile property test was made on the basis of ASTM D-638, the results of which are shown in Table 1.

Flame retarding agent (A)



polytribromostyrene

n = 50

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Table I

Compositions				Tensile property		UL94 burning property		Thermal stability (Tensile strength maintaining rate %)		Minimum injection pressure kg/cm <sup>2</sup> G	
PET (parts)	GF (parts)	SBO addition amount (parts)	Epoxy compound addition amount (parts)	Tensile strength (kg/cm <sup>2</sup> )	Elongation at break (%)	1/32"	1/8"	V-2	V-2	92	12
100	45	7	3	0.2	1410	3.1					
100	45	14	5	0.5	1350	3.7	V-2	V-0	92		14
100	45	16	6	0.5	1330	3.5	V-0	V-0	90		14
100	45	16	6	1.0	1410	3.8	V-0	V-0	93		18
100	45	18	7	0.5	1115	3.1	V-0	V-0	87		14
100	45	21	9	0.5	1070	3.0	V-0	V-0	85		13
EXAMPLES											
100	45	0	0	0	1460	3.7	HB	HB	79	10	
100	45	16	6	0	1210	2.6	V-0	V-0	77	7	
100	45	18	0	0	1110	3.5	V-2	V-2	75	8	

From the results of Table 1, it is clear that the composition according to the present invention has an excellent flame retardancy, and is, moreover, superior in the balancing of mechanical properties and fluidity.

EXAMPLE 2:

35 parts by weight of glass fibers, 12 parts by weight of the flame retarding agent (A) as employed in EXAMPLE 1, 5 parts by weight of antimony trioxide, and 1 part by weight of barium stearate were mixed with 100 parts by weight of polyethylene terephthalate (PET) with a relative viscosity of 1.35, with further addition thereto of various epoxy compounds as shown in Table 2 at the rate given in Table 2.

For the blending, the polyethylene terephthalate, flame retarding agent (A), antimony trioxide, barium stearate, and epoxy compounds were mixed by the V-blender, with subsequent addition thereto of the glass fibers.

The mixture thus obtained were subjected to melting and kneading at 280°C with the use of the 65 mmφ extruder to be formed into pellets. The resultant pellets were subjected to the injection molding by the method of EXAMPLE 1 after drying to prepare the burning test pieces and dumbbell test pieces, which were measured for physical properties.

The result of the measurements are shown in Table 2.

Table 2

Epoxy compound (added amount)	Tensile properties		UL94 flammability 1/32"
	Tensile strength (kg/cm <sup>2</sup> )	Elongation at break (%)	
EPIKOTE 815 (0.5)	1380	3.6	V-0
EPIKOTE 190 (made by Shell Company)	1490	4.1	V-0
(Tetrahydronaphthalic acid diglycidyl ester) (0.5)			
CARDURA E (made by Shell Company)	1320	3.4	V-0
{Undecanoic carboxylic acid monoglycidyl ester) (0.5)			
Ethylene-glycidyl- methacrylate copolymer (3.0)	1510	3.6	V-0
None	1190	3.2	V-0

From the results of Table 2, it is clear that the composition of the present invention is well balanced in the flammability and mechanical properties.

#### EXAMPLE 3

15 parts by weight of the flame retarding agent (A) as employed in EXAMPLE 1, 6 parts by weight of antimony trioxide (SBO), 0.1 part by weight of di-stearyl pentaerythritol diphosphite (trade name HI-MO, made by Sanko Kagaku) and 0.5 part by weight of "EPIKOTE 190" were mixed with 100 parts by weight of polybutylene terephthalate having a relative viscosity at 1.48, and 35 parts by weight of glass fibers.

For the blending, polybutylene terephthalate pellets and flame retarding agent (A), antimony trioxide, di-stearyl-pentaerythritol diphosphite, and "EPIKOTE190" were first mixed by the V-blender preliminarily, and the glass fibers were mixed, with attention directed so as not to excessively open said glass fibers.

The resultant mixture was melted and kneaded at 250°C by the 65mm<sup>φ</sup> extruder so as to be formed into pellets. The pellets thus obtained were formed after drying, into the burning test pieces (1/32") and dumbbell test pieces at a molding temperature of 250°C and a mold temperature of 80°C.

Upon measurement of the flammability and tensile properties in the similar manner as in EXAMPLE 1, the tensile strength of 1360 kg/cm<sup>2</sup> and UL94 flammability of V-0, were obtained, with the minimum injection pressure of 17 kg/cm<sup>2</sup>G and thermal stability of 86%.

Meanwhile, as a result of similar tests made on the composition to which only "EPIKOTE190" was not added, the flammability was V-0, but the tensile strength was reduced to 1210 kg/cm<sup>2</sup>, with simultaneous decrease of the minimum injection pressure to 9 kg/cm<sup>2</sup>G and that of the thermal stability to 73%.

#### EXAMPLE 4

100 parts by weight of polybutylene terephthalate with a relative viscosity of 1.70, 17 parts by weight of the flame retarding agent (A) employed in EXAMPLE 1, 7 parts by weight of antimony trioxide, 0.1 part by weight of thiadistearylpropionate (trade name "Sumilizer TPS", made by Sumitomo Kagaku), and 0.6

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parts by weight of "CARDURA E" as epoxy compound were mixed at the above rate.

For the blending, the V-blender were employed, and the resultant mixture was melted and kneaded at 250°C with the use of a 30 mmØ extruder to be formed into pellets. After having been dried in hot air at 130°C for 5 hours, the pellets were formed into the burning test pieces (1/8") and dumbbell test pieces at a molding temperature of 250°C and a mold temperature of 80°C. Upon subjecting the test pieces to the tensile property tests and UL94 flammability tests in the similar manner as in EXAMPLE 1, the tensile strength of 580 kg/cm<sup>2</sup>, elongation at break of 40% and UL94 flammability of V-0 were obtained.

When the physical property tests were made on the composition without addition of "CARDURA E" in the similar manner, the flammability was still V-0, but the tensile strength was lowered to 560 kg/cm<sup>2</sup>, with simultaneous reduction of the elongation at break to 20%.

What is claimed is:

1. A flame retarding polyester composition which comprises (a) 3 to 45 parts by weight of a halogen containing polystyrene and/or a halogen containing poly- $\alpha$ -methylstyrene represented by formula (I) given below, (b) 1 to 20 parts by weight of antimony trioxide, and (c) 0.1 to 10 parts by weight of epoxy compound which are mixed with 100 parts by weight of thermoplastic polyester,



Wherein R represents a hydrogen atom or methyl group, X represents a bromine or chlorine atom, p is an integer of 1 to 5, and n is an integer larger than 2.

2. A flame retarding polyester composition as claimed in Claim 1, wherein said thermoplastic polyester is a polyethylene terephthalate.
3. A flame retarding polyester composition as claimed in Claim 1, wherein said thermoplastic polyester is a polybutylene terephthalate.
4. A flame retarding polyester composition as claimed in Claim 1, wherein said halogen containing polystyrene is a polytribromostyrene.

5. A flame retarding polyester composition as claimed in Claim 1 or 4, wherein the total amount of halogen containing polystyrene and/or halogen containing poly- $\alpha$ -methylstyrene is in the range of 5 to 40 parts by weight.
6. A flame retarding polyester composition as claimed in Claim 1, wherein the epoxy compound is glycidyl esters of aliphatic acids or cycloaliphatic carboxylic acids.
7. A flame retarding polyester composition as claimed in Claim 1, wherein the epoxy compound is bisglycidyl ether of bisphenol A.
8. A flame retarding polyester composition as claimed in Claim 1, wherein said epoxy compound is ethylene/glycidyl methacrylate copolymer.
9. A flame retarding polyester composition as claimed in Claim 1, 6, 7 or 8, wherein the amount of addition of the epoxy compound is in the range of 0.2 to 9 parts by weight.
10. A flame retarding polyester composition as claimed in Claim 1, further containing fibrous and/or granular reinforcement.
11. A flame retarding polyester composition as claimed in Claim 10, wherein said reinforcement is glass fibers.



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EUROPEAN SEARCH REPORT

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Application number  
EP 81 10 7495

DOCUMENTS CONSIDERED TO BE RELEVANT			CLASSIFICATION OF THE APPLICATION (Int. CL <sup>3</sup> )
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	
X	<u>FR - A - 2 367 126 (B.A.S.E.)</u> * claims 1-6 * & GB - A - 1 588 642 --- <u>FR - A - 2 254 606 (CIBA)</u> * claims 1-12 * & GB - A - 1 467 523 -----	1-5,10 -11	C 08 L 67/02
		1-5,10 -11	TECHNICAL FIELDS SEARCHED (Int. CL <sup>3</sup> )  C 08 L 67/02 67/00
			CATEGORY OF CITED DOCUMENTS  X: particularly relevant A: technological background O: non-written disclosure P: intermediate document T: theory or principle underlying the invention E: conflicting application D: document cited in the application L: citation for other reasons
	<input checked="" type="checkbox"/> The present search report has been drawn up for all claims		&: member of the same patent family. corresponding document
Place of search	The Hague	Date of completion of the search	Examiner DECOCKER